

Model-mapped RPA for Determining the Effective Coulomb Interaction

Hirofumi Sakakibara^{1,2,*}, Seung Woo Jang³, Hiori Kino⁴, Myung Joon Han³, Kazuhiko Kuroki⁵, and Takao Kotani¹

¹*Department of applied mathematics and physics, Tottori university, Tottori 680-8552, Japan*

²*Computational Condensed Matter Physics Laboratory, RIKEN, Wako, Saitama 351-0198, Japan*

³*Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, Korea*

⁴*Research Organization of Science and Technology, Ritsumeikan university, Kusatsu, Shiga 525-8577, Japan*

⁵*National Institute for Materials Science (NIMS), Sengen 1-2-1, Tsukuba, Ibaraki 305-0047, Japan and*

⁵*Department of Physics, Osaka University, Machikaneyama-Cho, Toyonaka, Osaka 560-0043, Japan*

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We present a new method to obtain a model Hamiltonian from first-principles calculations. The effective interaction contained in the model is determined on the basis of random phase approximation (RPA). In contrast to previous methods such as projected RPA and constrained RPA (cRPA), the new method named “model-mapped RPA” takes into account the long-range part of the polarization effect to determine the effective interaction in the model. After discussing the problems of cRPA, we present the formulation of the model-mapped RPA, together with a numerical test for the single-band Hubbard model of $\text{HgBa}_2\text{CuO}_4$.

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I. INTRODUCTION

Recently, we often treat low-temperature physical phenomena in correlated materials by a two-step procedure, that is, deriving a model Hamiltonian from a first-principles calculation and solve the model Hamiltonian¹⁻⁴. Thus, the procedure contains two key points:

(A) How to derive a model Hamiltonian.

(B) How to solve the model Hamiltonian.

For step (B), we can use various many-body calculation techniques to solve the model Hamiltonian. The techniques for step (B) can be dynamical mean-field theory (DMFT)⁵, quantum Monte Carlo (QMC)⁶ methods, and so on. The two-step procedure is generally applicable to strongly correlated systems such as high- T_c superconductors and metal-insulator transitions.

In this paper, we focus on step (A), that is, how to derive a model Hamiltonian from first-principles calculations, especially the effective interaction contained in the Hamiltonian (we neglect phonons here). If step (A) is well established and combined with a reasonable technique in step (B), we can even evaluate the transition temperature of superconductivity T_c for given crystal structures without introducing parameters by hand⁷⁻¹³. This means we can use the two-step procedure for material informatics combined with databases of crystal structures. In future, we may find new high- T_c superconductors among thousands of possible candidates^{14,15} in the two-step procedure.

Let us present an overview of step (A). We have various first-principles methods to determine one-body Hamiltonian H^0 . These method are the local density approximation (LDA), the quasiparticle self-consistent GW method (QSGW)¹⁶⁻¹⁹, and so on. The one-body Hamiltonian H^0 describes an independent particle picture. The static screened Coulomb interaction $W(\mathbf{r}, \mathbf{r}', \omega = 0)$ can be calculated in the random-phase approximation (RPA). From H^0 , we can construct a set of the atomic-like localized orbitals $\{\phi_{Ri}(\mathbf{r})\}$ which describe

the low-energy bands. The orbitals can be constructed, for example, by the method of the maximally localized Wannier functions²⁰. The orbitals span a model Hilbert space \mathcal{M} . The choice of \mathcal{M} is not unique and has ambiguity. If necessary, we should use a larger \mathcal{M} to reduce the ambiguity. However, step (B) requires a sufficiently small \mathcal{M} to ensure tractability within current computational resources. The one-body part of the model Hamiltonian in \mathcal{M} can be determined by the projection of H^0 into \mathcal{M} . As for the effective interaction, we cannot simply project $W(\mathbf{r}, \mathbf{r}', \omega = 0)$ into \mathcal{M} . In advance, we have to remove the screening effect expected within the model. This is necessary to avoid double counting of the screening effect. This idea was first introduced in the projected RPA (pRPA) method by Kotani²¹. This was followed by the constrained RPA (cRPA) by Aryasetiawan *et al.*²². cRPA has been applied to several compounds to analyze strongly correlated systems²³⁻²⁸. Miyake implemented a Wannier-based modified cRPA in the `ecalj` package²⁹. Şaşoğlu, Friedrich, and Blügel also proposed a modified cRPA applicable to cases with entangled bands³⁰. However, cRPA contain theoretical problems as we discuss in Sec. II.

The reliability of the model Hamiltonian obtained in step (A) is determined by the reliability of the first-principles calculation. Most popular calculations are in LDA. However, LDA often gives an unreliable independent particle picture, especially for transition metal oxides and f -electron materials. A well-known problem is the underestimation of the band gaps. Furthermore, there are problems with the bandwidth, the positions of the $3d$ bands and $4f$ bands relative to the oxygen bands, and so on. In such cases, we need to use advanced methods such as hybrid functional methods³¹ or QSGW¹⁶⁻¹⁹. One of the advantages of QSGW is that H^0 and $W(\mathbf{r}, \mathbf{r}', \omega = 0)$ are determined simultaneously in a self-consistent manner. QSGW has even been applied to metallic ground states. For example, Han *et al.* recently applied QSGW to $\text{LaNiO}_3/\text{LaAlO}_3$ ³², Jang *et al.* applied QSGW to high- T_c materials³³, and Ryee *et al.* applied QSGW to SrRuO_3 and Sr_2RuO_4 ³⁴. To handle such metallic systems, QSGW is more reliable than the hybrid functional methods³⁵.

Furthermore, Deguchi, Sato, Kino, and Kotani have recently shown that a QSGW-based hybrid method can systematically give a good description for a wide range of materials³⁶. These calculations were performed by the first-principles calculation package `ecalj`³⁷, which is based on a mixed-basis full-potential method, the linearized augmented plane wave, and muffin-tin orbital method (the PMT method)^{38–40}. It is freely available from github³⁷.

In this paper, we propose a new method named model-mapped RPA (mRPA). This is based on an assumption of the existence of a model Hamiltonian that explains the low-energy physical properties of materials. This assumption is standard in the field of model calculations. For example, we may assume that low-energy physical properties can be quantitatively understood by a Hubbard model. Then, the role of mRPA is to determine the interaction parameters in the model.

After we review cRPA and point out its problems in Sec. II, we give a formulation of mRPA in Sec. III. Then we show how it works for a test case of single-band Hubbard model for the high- T_c superconductor $\text{HgBa}_2\text{CuO}_4$ in Sec. IV, followed by a summary.

II. CRPA AND ITS PROBLEMS

In the first-principles calculations, the screened Coulomb interaction $W(\mathbf{r}, \mathbf{r}', \omega)$ in RPA is given by

$$W = \frac{1}{1 - vP}v, \quad (1)$$

where $v(\mathbf{r}, \mathbf{r}')$ and $P(\mathbf{r}, \mathbf{r}', \omega)$ are the Coulomb interaction and the non-interacting proper polarization, respectively. $\frac{1}{X}$ (written as X^{-1} below) denotes the inverse of matrix X . P consists of a product of two Green functions $G_0 = 1/(\omega - H^0)$. We can represent the quantities W , v , and P expanded in an improved version of the mixed product basis (MPB). The MPB was originally introduced by Kotani in Ref. 41. Then, the MPB was improved by Friedrich, Blügel, and Schindlmayr⁴². We usually use the improved MPB.

Let us recall the idea of the so-called cRPA. We first choose a model space \mathcal{M} spanned by a basis set of atomic-like localized orbitals, $\{\phi_{Ri}(\mathbf{r})\}$, where R is the index of the primitive cell and i is the index used to specify an orbital in the cell. In the following, we use the notations, $1 \equiv n_1 \equiv R_1 i_1$ and

$$(1, 2|W|2', 1') = (\phi_1, \phi_2|W|\phi_{2'}, \phi_{1'}) \\ \equiv \int d^3r d^3r' \phi_1^*(\mathbf{r}) \phi_{2'}^*(\mathbf{r}') W(\mathbf{r}, \mathbf{r}', \omega) \phi_{2'}(\mathbf{r}') \phi_1(\mathbf{r}), \quad (2)$$

as in the manner of Ref. 43. The eigenfunctions in \mathcal{M} are calculated from the Hamiltonian $H_M^0 \equiv \langle \phi_{Ri} | H^0 | \phi_{R'i'} \rangle$, that is, H_M^0 is the same as H^0 but restricted within the space \mathcal{M} . Then, we have the non-interacting proper polarization function $P_m(\mathbf{r}, \mathbf{r}', \omega)$ in \mathcal{M} . Let us consider the RPA-screened Coulomb interaction $W_m(\mathbf{r}, \mathbf{r}', \omega)$ in \mathcal{M} . It can be written as

$$W_m = \frac{1}{1 - U_m P_m} U_m, \quad (3)$$

where $U_m(\mathbf{r}, \mathbf{r}', \omega)$ is the (not yet determined) effective interaction between quasi-particles in \mathcal{M} . cRPA determines $U_m(\mathbf{r}, \mathbf{r}', \omega)$ by assuming

$$W_m = W, \quad (4)$$

with Eq. (3). From Eqs. (1), (3), and (4), we have

$$U_m = \frac{1}{1 - v(P - P_m)}v, \quad (5)$$

that is, we can calculate U_m from v , P , and P_m . Then, we calculate the on-site interaction U for the model as

$$U \equiv U_{n,n,n,n} = (\phi_n, \phi_n | U_m | \phi_n, \phi_n). \quad (6)$$

If necessary, we can calculate any elements of the interaction given as $U_{1,2,2',1'} = (\phi_1, \phi_2 | U_m | \phi_{2'}, \phi_{1'})$. However, in the usual cRPA, we only calculate the set of parameters $\{U\}$ used by the model. In summary, for the choice of a localized basis set $\{\phi_{Ri}\}$, we determine a set of interactions $\{U\}$ of the model in cRPA, where U are ω -dependent.

The cRPA, which appears to be reasonable, however, contains the following three problems.

(i) Range truncation problem

$W = W_m$ is satisfied only when we take all possible elements of $U_{1,2,2',1'}$ in cRPA. However, practically adopted models consider a limited number of U . Note that U_m given in Eq. (5) is inevitably long-range. This is because the strong screening effects such as metallic screening contained in P_m are removed from the total polarization P . This problem is well illustrated when only the on-site U is used. In this case, because we use only the on-site part of U_m evaluated from the right-hand side of Eq. (5), W_m given in Eq. (3) cannot satisfy the condition $W_m = W$.

Schüler *et al.*⁴⁴ proposed a method to solve an extended Hubbard model with non local interaction. However, the method is not applicable to the long-range interaction $\propto 1/|\mathbf{r} - \mathbf{r}'|$ without modification. Hansmann *et al.*⁴⁵ calculated the long-range behavior of the effective interaction $\propto 1/|\mathbf{r} - \mathbf{r}'|$ by cRPA, and presented a method to solve a model Hamiltonian taking into account the long-range interaction. In contrast, we can handle the same problem using a model Hamiltonian with the short-range interaction given by mRPA. This is because mRPA downfolds the long-range interaction into the short-range interaction as described in Sec. III.

(ii) Positive definiteness and causality problem

$-(P - P_m)$ in the denominator of Eq. (5) should be positive definite at $\omega = 0$. If this is not satisfied, we obtain unphysical results for U_m having eigenvalues larger than the bare interactions v . Furthermore, the imaginary (anti-hermitian) part of $-(P - P_m)$ at any ω should be positive definite so as to satisfy causality. In the original idea of cRPA, $P - P_m$ does not necessarily satisfy this condition for the case of entangled bands. For this reason, Kotani avoided the idea of cRPA and

proposed pRPA, which satisfies the above conditions²¹. Recently, two other procedures satisfying the conditions have been proposed with a modification of the definition of $P - P_m$ in the cRPA. One given by Miyake, Aryasetiawan, and Imada, neglects the off-diagonal elements between \mathcal{M} and residual space in the one-body Hamiltonian [see Fig. 1 and Eq. (8) in Ref. 29]. Thus, the condition is clearly satisfied. The other is given by Şaşıoğlu, Friedrich, and Blügel, where a projection procedure of eigenfunctions to \mathcal{M} is used to satisfy the condition³⁰. Strictly speaking, neither procedure should be identified as cRPA, since the key advantage of cRPA, Eq. (4), is no longer satisfied.

Note the generality of the causality problem. For example, in the GW+DMFT formulation^{46,47} as an extension of the LDA+DMFT^{48–50}, the on-site part of the GW self-energy is simply substituted with the DMFT self-energy. Then, we may have a causality problem if we have a GW self-energy whose imaginary part is larger than that of the DMFT self-energy.

(iii) energy window problem

In Table I, we have calculated the static (at $\omega = 0$) part of W and U for the paramagnetic Ni, where we use the cRPA method given by Şaşıoğlu, Friedrich, and Blügel⁵¹. We considered two cases for the energy window; the narrower one is $-8 \sim 1$ eV and the wider is $-10 \sim 10$ eV. In contrast to the small difference in W for the different energy window, we see a large difference in U . The value of 3.56 eV is in good agreement with that in Ref. 30. As shown in Fig. 5 in Ref. 51, they used such a wide energy window. The value of 2.25 eV for the narrower window is significantly different from 3.56 eV. This difference is because of the difference in P_m , which describes the polarization of the 3d electrons. In the case of wider windows, we remove more polarization, resulting in larger values of U . This results in an inevitable ambiguity in the cRPA because we have almost the same energy bands (the same eigenvalue dispersions in the Brillouin zone) for both windows. In addition, we have no definite criteria for choosing a certain energy window.

We expect that a similar ambiguity also exists in other versions of cRPA. Miyake, Aryasetiawan, and Imada, successfully obtained flat low-energy behaviors, as shown in Fig. 3 of Ref. 29, similar to that in Fig. 1 of Ref. 21. However, the procedure of neglecting the off-diagonal elements (equivalent to how to choose the \mathcal{M}) is ambiguous. From Fig. 3 in Ref. 29, we guess that the ambiguity of $U(\omega = 0)$ in their method can be ≈ 1 eV [from the degree of freedom in the choice of the energy window and \mathcal{M} , we may have various possible extrapolations of $U(\omega)$ to $\omega = 0$].

Although the new method, the mRPA, formulated in Sec. III can remedy these problems, we need to pay attention to the inevitable limitations of model Hamiltonians including no long-range interactions. Recall that plasmons (charge fluctuations) do not satisfy the Goldstone's theorem because of the

TABLE I. Static screened Coulomb interactions W and U in cRPA method³⁰ for two different outer energy windows, $-8 \sim 1$ eV, and $-10 \sim 10$ eV for paramagnetic Ni. For both energy windows, we have almost the same 3d bands for the model space \mathcal{M} . We use $12 \times 12 \times 12$ \mathbf{k} points in the Brillouin zone in the tetrahedron method^{16,17}. There is a large difference between the two U values. See text.

	-8~1 eV	-10~10 eV
$W[\text{eV}]$	1.19	1.40
$U[\text{eV}]$	2.25	3.56

$1/r$ behavior of the Coulomb interaction. Such model Hamiltonians cannot describe this correctly. The long-range limit of longitudinal spin fluctuations, as well. Model Hamiltonians can only be justified when these problems are irrelevant.

III. FORMULATION OF THE MRPA

Let us assume that a model Hamiltonian H_M in the model space \mathcal{M} can describe low-energy excitations very well. Here we formulate mRPA, which determines the parameters included in H_M . H_M is given as

$$H_M = H_M^0 + U_M - \bar{U}_M, \quad (7)$$

where H_M^0 is the one-body Hamiltonian, obtained from a first-principles method such as QSGW. U_M is the spin-independent effective interaction specified by a set of parameters $\{U\}$ (here we do not consider the ω -dependence of these parameters). The terms $H_M^0 + U_M$ are those in Eq. (1) in Ref. 43. \bar{U}_M is the one-body counter term so that the effect of $U_M - \bar{U}_M$ is canceled out when we apply the first-principles method to the model described by H_M ⁵². In the U_M , used elements $U_M(1, 2, 2', 1')$ are given by a set of a finite number of parameters $\{U\}$. We usually allow only the short-range terms; for example, we only allow the on-site terms in the case of the Hubbard model.

Let us explain how to determine $\{U\}$ (or U_M equivalently) in mRPA. If we apply RPA to the model Hamiltonian H_M , we have the screened Coulomb interaction of the model $W_M(1, 2, 2', 1')$ as

$$W_M = \frac{1}{1 - U_M P_M} U_M, \quad (8)$$

where we use the proper non-interacting polarization P_M calculated from H_M^0 . In mRPA, we only consider the case at $\omega = 0$ in Eqs. (8)–(11).

Note the difference between Eq. (3)(cRPA) and Eq. (8)(mRPA). In Eq. (3), U_m inevitably become long-range as $\propto 1/r$, while U_M is short-range such as on-site only in Hubbard model. That is, P_M , U_M and W_M are non-zero just on the limited number of discrete index set of \mathcal{M} in Eq. (8).

For the theoretical correspondence, we require W_M to satisfy

$$W_M(1, 2, 2', 1') = W_{FP}(1, 2, 2', 1'), \quad (9)$$

in mRPA in order to determine U_M . Here, $W_{FP}(1, 2, 2', 1') \equiv (1, 2|W|2', 1')$ is the quantity calculated from W in the first-principles method using Eq. (1). It is not possible to satisfy

Eq. (9) for all the matrix elements of $W_M(1, 2, 2', 1')$; we satisfy a subset of Eq. (9) corresponding to the degree of freedom of $U(1, 2, 2', 1')$ used in U_M of Eq. (7). Thus, the subset of Eq. (9) can determine $\{U\}$ uniquely. Then, we can determine U_M from Eq. (8) so as to satisfy

$$\frac{1}{1 - U_M P_M} U_M = W_{FP}, \quad (10)$$

or

$$U_M = \frac{1}{1 + W_{FP} P_M} W_{FP}, \quad (11)$$

equivalently.

By definition, mRPA satisfies Eq. (9) exactly, where $W_M(1, 2, 2', 1')$ is expressed in terms of U_M and P_M in Eq. (8). This is in contrast to the case of cRPA, which can not usually satisfy Eq. (4) since cRPA usually discards the off-site part of U_m . Thus, we are free from the problem (i) in Sec. II in the case of mRPA.

We may have cases that U_M satisfying Eq. (9) cannot be found. This is because W_M has the upper limit $-1/P_M$ (\approx the bandwidth of H_M^0); for $U_M \rightarrow \infty$, we have $-1/P_M$ can be seen from Eq. (8). Thus, we cannot determine U_M for very large W_{FP} . This can be clearly seen in Fig. 1 as explained later. In such cases, we need to use a larger M . This is not an intrinsic problem of mRPA but a problem associated with choosing of too small M .

The causality problem (ii) in Sec. II does not arise, since ω -dependence of U_M is meaningless in mRPA; we use the condition given by Eq. (9) only at $\omega = 0$. In our opinion, we rather have to use a larger M for better results, instead of taking the ω -dependence into account in theoretical treatments. If we take the ω -dependence of the effective interaction correctly, we inevitably have to treat a quantum Langevin equation with electron thermal bath. Such a treatment is far beyond our current numerical techniques because it requires an enormous computational effort.

Let us consider problem (iii) in Sec. II in the case of mRPA. In mRPA, P_M is only determined from the energy bands of H_M^0 . The choice of the Wannier functions (the choice of M) can slightly change W_{FP} . This yields the slight ambiguity of U_M via Eq. (10). This is inevitable as long as we derive a model from first-principles calculations. In contrast, cRPA has further ambiguity in the polarization of P_m in Eq. (5) owing to the ambiguity of the choice of Wannier functions as we have shown in Table I.

IV. NUMERICAL TEST FOR A SINGLE BAND HUBBARD MODEL OF HGBA₂CUO₄

Here we present a test calculation to see how mRPA works in comparison with cRPA. We take a single-band Hubbard model for stoichiometric HGBa₂CuO₄. We treat two cases where H_M^0 is determined by LDA or by QSGW. The space M is chosen by a procedure based on maximally localized Wannier functions⁵³. The term \tilde{U}_M in Eq. (7) is irrelevant in the single-band case since it gives a constant potential shift. As

TABLE II. Calculated values of U_M (mRPA) and U_m (cRPA) for a single-band model of HGBa₂CuO₄, together with W_{FP} . Note that we only consider the values at $\omega = 0$. We use the tetrahedron method in Ref. 16 for the evaluation of U_M and W_{FP} , where we use $8 \times 8 \times 4$ \mathbf{k} points in the Brillouin zone, as was used in Ref. 54. The values of U_m in cRPA³⁰ are the same as those presented in our previous paper⁵⁵. U_M is determined by mRPA as illustrated in Fig. 1.

	W_{FP} [eV]	U_M [eV]	U_m [eV]
QSGW	0.67	5.2	3.7
LDA	0.85	3.9	2.0

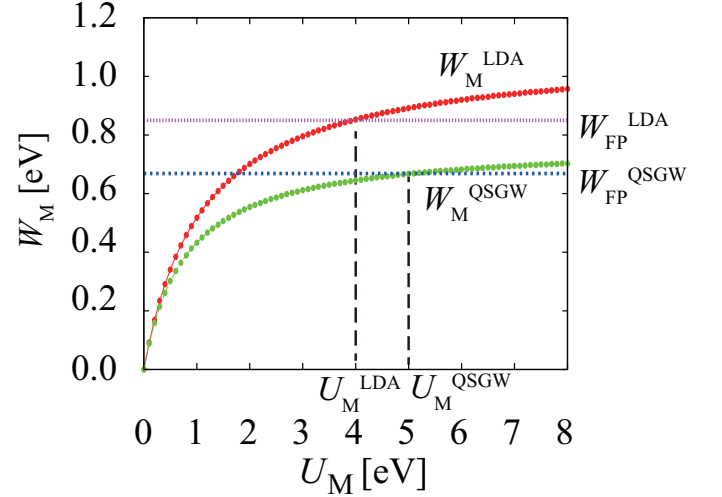


FIG. 1. (Color online) Calculated W_M as a function of U_M in Eq. (8) for a single-band model of HGBa₂CuO₄. The Green line represents QSGW, the red line represents LDA. The values of U_M in mRPA are obtained at the intersections between the curves of W_M and the horizontal lines of W_{FP} .

we use the single-band Hubbard model, we obtain U_M and W_M as scalars.

The two curves in Fig. 1 show W_M as functions of U_M given by Eq. (8), where we use P_M calculated from H_M^0 by QSGW or by LDA. As a function of U_M , these curves are initially linear near $U_M = 0$ and saturate toward $\frac{1}{P_M}$. The difference between the two curves is due to the size of $-1/P_M$ corresponding to the size of the bandwidth³³. The two horizontal lines show the values of W_{FP} (0.67 and 0.85 eV) calculated by the first-principles RPA method as shown in Table II. Using the condition Eq. (9), we can determine U_M for QSGW and for LDA as illustrated in Fig. 1.

The obtained values of U_M are shown in Table II, together with the cRPA values U_m obtained by the method in Ref. 30. The values of U_M are 1.5 ~ 2.0 eV larger than those of U_m . This is because we use the on-site interaction only in the present model. If we take into account off-site interactions, U_M will be reduced. In other words, mRPA downfolds the off-site interactions into the on-site interaction.

In Fig. 1, we see that the determined values of U_M are sensitive to the values of W_{FP}^{LDA} and W_{FP}^{QSGW} . This is because the calculated values of U_M are close to the upper limit of RPA, $-1/P_M$ at $U_M \rightarrow \infty$. The derivatives dW/dU at U_M are

rather small, 0.060 for LDA and 0.019 for QSGW. This sensitivity may indicate the lack of suitability (or limitation) of the single-band Hubbard models for $\text{HgBa}_2\text{CuO}_4$. If we use a larger M , we will be able to avoid such cases of $U_M \sim -1/P_M$.

V. SUMMARY

We have presented mRPA to determine model Hamiltonians based on first-principles calculations. mRPA is formulated starting from the assumption of the existence of a model Hamiltonian that explains the low-energy physical properties of materials. Then we determine the effective interactions contained in the Hamiltonian by matching the first-principles RPA calculations and the RPA calculations using the model Hamiltonian. mRPA is free from the theoretical problems in cRPA, which are discussed in Sec. II. Thus, mRPA is less ambiguous and logically clearer than cRPA. Through the model

Hamiltonian obtained by mRPA, we will be able to predict the critical temperatures of superconductors based on first-principles calculations.

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- * sakakibara.tottori.u@gmail.com
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